Fuel 115 (2014) 54–61

Contents lists available at SciVerse ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

# Air and air-steam gasification of sewage sludge. The influence of dolomite and throughput in tar production and composition

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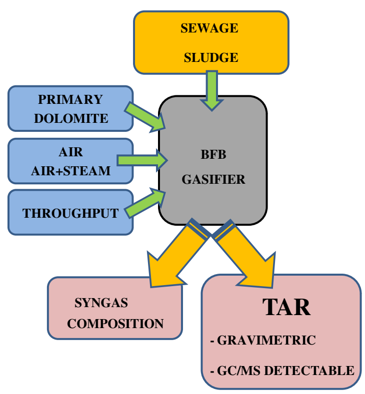
## HIGHLIGHTS

- The effect of some primary measures on sewage sludge gasification products was tested.
- Higher throughputs decreased the $H_2$ content of the syngas and increased tar production.
- The use of dolomite showed gravimetric tar removal efficiencies of up to 71%.
- Dolomite performance remained fairly constant over the range of studied throughputs.
- Under the tested experimental conditions, syngas dew point never dropped below 110 °C.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

*Article history:*
Received 30 May 2013
Received in revised form 1 July 2013
Accepted 1 July 2013
Available online 15 July 2013

*Keywords:*
Sewage sludge gasification
Primary measures
Gravimetric tar production
GC-detectable tar

## ABSTRACT

The influence of throughput (TR), steam and the use of dolomite (as primary catalyst) over the sewage sludge gasification products was investigated. For this purpose, experiments were conducted in an atmospheric fluidised bed reactor using air and air + steam as gasifying agents. The analysis of the results was mainly focussed on the gas composition, the gravimetric tar production, and the GC-detectable tar composition (and dew point estimations). According to the obtained results, higher TRs decreased the $H_2$ content of the produced gas and clearly increased the gravimetric tar production. The use of air + steam, especially in the presence of dolomite, increased the $H_2$ content (between 20% and 36%) and decreased the gravimetric tar production over all the tested TR, reaching tar removal efficiencies of up to 71%. Regarding the GC-detectable tar, higher TRs increased the heavy polyaromatic hydrocarbons production, steam slightly increased the water soluble tar content while the use of dolomite decreased the yield of all the tar classes except light aromatic hydrocarbons. Under the tested gasification conditions, the gas dew point never dropped below 110 °C, value far above the recommended temperature when the syngas is to be used for engine applications.

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## 1. Introduction

Waste management is a worldwide environmental problem that can be partially solved by recovery technologies which are able to obtain energy from waste. Among the different Waste-to-Energy (WtE) options currently available, gasification is proposed as an alternative to conventional combustion plants [1]. This technology has the advantages of incineration, such as destruction of pathogenic bacteria and volume reduction, as well as the additional benefits of higher energy recoveries and lower-cost atmospheric emissions control [2].

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http://dx.doi.org/10.1016/j.fuel.2013.07.003

In this context, there is an increasing interest in the gasification of sewage sludge [3,4]. Sewage sludge is the liquid or semi-liquid waste generated in wastewater treatment plants. The physical and chemical properties of this waste make it an interesting feedstock for gasification applications [5].

The main product obtained from sewage sludge gasification is a combustible gas (syngas), basically composed of $H_2$, CO, $CO_2$, $CH_4$ and $N_2$ (in gasification with air). The syngas has many applications such as heat/electricity production, chemical synthesis and fuel cells. However, most of these applications need extensive and costly gas cleaning systems to remove the impurities it contains [6]. Special attention must be directed to its tar content, which can cause serious operational interruptions that clearly limit the use of syngas [7].

The gasification conditions greatly influence the tar production and syngas composition. Parameters such as temperature, residence time, the gasifying agent used and the utilisation of catalysts have a large effect on the gasification products [8]. Regarding the gasifying agents, the most widely used are air, oxygen, steam or a mixture of these [9,10]. However, temperature and residence time highly depend on the gasification system used. For instance, temperatures between 800 and 900 °C are typical in atmospheric air-blown bubbling fluidised bed gasifiers whereas temperatures of up to 1100 °C or even 1500 °C are typical for fixed bed and entrained flow gasifiers [11,12].

Regarding the use of catalysts in gasification processes, two different configurations are feasible. Catalysts can be located inside the gasifier (primary methods) or downstream the gasifier (secondary methods). Although the secondary methods have proven their effectiveness for tar removal and syngas conditioning [13–16], they are either costly or complex for small or medium scale systems [17,6].

Considering this, a large amount of research has been made to study the effect of different primary catalysts on the gasification products [18,19]. Olivine, dolomite and alumina are the most common primary catalysts used in the biomass and sewage sludge gasification [20–23]. Among the different available options, dolomite is reputed as the most efficient catalyst in improving gasification performance and reducing the tar production. Despite the fact that this cheap catalyst has clear advantages, dolomite suffers from mechanical attrition in fluidisation environments, which can affect its performance [24]. More effective nickel-based catalysts improve tar reduction when are used as in-bed material, but their rapid degradation and the fact that they can contaminate the ashes make this option unfeasible so far [25].

At this point, it is important to highlight that catalyst performance can be greatly influenced by throughput (TR, hereinafter), defined as the kilograms of sewage sludge "as received" fed to the gasifier per hour and per square metre of cross sectional area of the gasifier. According to [8], some studies developed at small scale use very low TR (soft conditions, TR close to 100–150 kg/h $m^2$). As a result of that, the tar removal efficiencies found may be very different from those obtained at commercial scale, with TR around 750 kg/h $m^2$. This assertion is based on the extensive Corella and co-workers's experience, who have carried out numerous studies on biomass gasification working under different TRs and using different kind of catalysts [20,26–30]. However, an effort is still needed to know the effect of the TR alone on catalysts performance due to the fact that most of the above-mentioned works were conducted under different gasification conditions, making difficult the analysis of the effect of the TR on the tar removal efficiency.

In addition, it is known that the final use of the produced gas defines the need for tar conversion [17]. However, regardless of the tar concentration, the nature of the tar is a key parameter for the assessment of the suitability of the syngas for a given application [25]. Tar composition determines the dew point of the syngas, which may limit its utilisation in power applications [6]. Taking this into account, the tar concentration is an interesting but incomplete information in the study of the syngas applicability.

Although there is much information on the effects of other parameters (e.g. temperature and gasifying agent) in the gasification performance, the objective of this paper is to provide insight on the effect of TR in the behaviour of primary dolomite and the resulting gasification products. The final aim is to gain knowledge about the expected tar removal efficiency under gasification conditions similar to those used at industrial scale. This was accomplished by analysing the changes found in gas composition, gravimetric tar production ($Y_{tar}$) and tar composition, in order to estimate the syngas dew point, which is an essential parameter to evaluate the suitability of the produced gas for power applications.

## 2. Materials and methods

### 2.1. Materials

The dried sludge samples were supplied by a sludge thermal drying plant located in Madrid, Spain, They were received as ball-shaped samples of 2–5 mm in diameter. The results of the proximate and ultimate analyses of the sludge samples are shown in Table 1. These data were used to estimate the low heating value (LHV) of the sludge (13.1 MJ/kg) by the means of the modified Dulonǵs formula [31]. The sludge was crushed and sieved to obtain samples with a particle size in the range of 250–500 μm. Silica sand was used as bed material and dolomite, supplied by Dolomitas del Norte SA, Cantabria (Spain), was used as catalyst. Both silica sand and dolomite were also sieved at the same particle size of the sludge samples.

### 2.2. Laboratory scale plant

Experiments were carried out in a laboratory scale plant (detailed information can be found in [21]). The reactor used was a stainless steel fluidised bed gasifier followed by a freeboard, both of them electrically heated. The fluidised bed gasifiers allow variations in fuel quality and the scaling-up of the process, making them ideal for the processing of biomass and wastes [6].

The bed height was kept at 100 mm by a concentric pipe which goes through the distributor plate and lets the overflowing material to be collected and stored in a discharge tank. The gasifying agent entering the reactor (air or air + steam) was electrically preheated.

A cyclone and a micronic filter were placed downstream of the freeboard (inside a hot box) to remove the particles from the syngas. Tar collection was carried out in a set of 6 impinger bottles containing isopropanol according to the European Committee for Standardization (CEN) tar protocol [32].

Finally, the gas production was measured by a mass flow metre while the dry gas composition ($N_2$, $O_2$, $H_2$, CO, $CO_2$, $CH_4$, $C_2H_6$ and $C_2H_4$) was determined using a microgas chromatograph (Micro-GC, Varian CP-4900).

### 2.3. Determination of the tar production and tar composition

The samples collected in the 6 impinger bottles were mixed thoroughly to assure homogeneity.

The gravimetric tar production was determined by distillation of the isopropanol-tar solution to remove the solvent (isopropanol). After the distillation step, the residue (tar) was dried at room temperature until constant weight. Finally, the sample was weighed [32].

**Table 1**
Elemental analysis of sewage sludge (dry basis) from a sewage sludge drying plant.

| Parameter | | Value[a] | Analytical method |
|---|---|---|---|
| Moisture (%) | | 8.7 | UNE-EN 12880-2001 |
| Organic mat. (%) | | 58.3 | UNE-EN 12879-2001 |
| Ash (%) | | 41.7 | UNE-EN 12879-2001 |
| Carbon (%) | | 29.5 | Elementary microanalyser LECO CHNS-932 |
| Nitrogen (%) | | 4.1 | |
| Hydrogen (%) | | 4,9 | |
| Sulphur (%) | | 1.6 | |
| Oxygen (%) | By difference | 15.0 | |
| Heavy metals (mg/Kg) | Cu | 382.2 | UNE-EN 13346-2001 |
| | Ni | 70.0 | |
| | Pb | 109.6 | |
| | Zn | 1926.2 | |
| | Cd | 3.1 | |
| | Cr | 117.3 | |

[a] Mean value of three analytical assays.

Regarding tar characterisation, the isopropanol-tar solution was analysed by gas chromatography/mass spectrometry (GC/MS) using a Perkin Elmer Clarus 600T GC/MS, fitted with a Elite 5MS capillary column (30 m × 0.25 mm i.d.). The GC oven temperature program used was: initial isothermal at 50 °C for 5 min, heating rate of 8 °C/min up to 325 °C, final temperature held for 15 min. Sample injections of 1 μL were made at 1:25 split with an injector temperature of 275 °C. The carrier gas was helium at a 1.0 mL/min flow rate.

The species detected by the GC/MS are presented in Table 2. The gas dew point of each tar sample was estimated by using the complete model available on the "Thersites" website [33], published by the Energy research Centre of the Netherlands (ECN).

### 2.4. Experimental conditions

A set of tests was carried out to know the effect of TR, steam and dolomite on the gasification products. In all the tests, the equivalence ratio (ER, defined as the ratio between the flow rate of air introduced into the gasifier and the stoichiometric flow rate of air required for complete combustion of the sludge) and the temperature were set at 0.3 and 800 °C, respectively. The before mentioned values are in the usual range for this kind of experiments [18,22].

The experiments can be divided into three groups depending on their main objective:

- Influence of throughput, TR. In these tests, the flow rate of the sludge fed to the gasifier was modified to work under TRs of 110, 215 or 322 kg/h $m^2$. As the ER must be kept at 0.3, the air flow rate introduced in the gasifier was modified and therefore the space residence time of the gas in the gasifier (*srt*, defined as the gasifier volume divided by the air volumetric flow rate) changed. The corresponding *srt* values were 7.5, 3.7 and 2.5 s, respectively. It can be seen how a higher TR corresponds to a lower *srt*.
- Influence of steam as gasifying agent. A set of tests was carried out at the three TRs previously defined (110, 215 or 322 kg/h $m^2$) and introducing a mixture of steam and air until a steam-to-biomass ratio (S/B) of 1 was reached (S/B, defined as the flow rate of steam fed to the reactor divided by the flow rate of sludge on a dry and ash free basis introduced therein).
- Influence of the catalyst. Additional tests were performed by adding dolomite to the gasifier (10% by weight within the fed sludge) at the three different TRs and at S/B ratios of 0 (tests with air) and 1 (tests with air + steam). This amount of catalyst was set according to previous experiences [21].

**Table 2**
Tar species quantified by GC/MS.

| Compound | Raw formula | MW[a] | Class[b] |
|---|---|---|---|
| Benzene | $C_6H_6$ | 78 | – |
| Pyridine | $C_5H_5N$ | 79 | 2 |
| Toluene | $C_6H_5CH_3$ | 92 | 3 |
| Phenol | $C_6H_5OH$ | 94 | 2 |
| Benzonitrile | $C_6H_5CN$ | 103 | 2 |
| Styrene | $C_6H_5CH{=}CH_2$ | 104 | 3 |
| Indene | $C_9H_8$ | 116 | 4 |
| Indole | $C_8H_7N$ | 117 | 2 |
| Naphthalene | $C_{10}H_8$ | 128 | 4 |
| Quinolin | $C_9H_7N$ | 129 | 3 |
| Acenaphtylene | $C_{12}H_8$ | 156 | 4 |
| Biphenyl | $C_6H_5C_6H_5$ | 154 | 4 |
| Acenaphtene | $C_{12}H_{10}$ | 154 | 4 |
| Fluorene | $C_{13}H_{10}$ | 166 | 4 |
| Carbazole | $C_{12}H_9N$ | 197 | 4 |
| Dibenzofuran | $C_{12}H_8O$ | 168 | 2 |
| Phenanthrene | $C_{14}H_{10}$ | 178 | 4 |
| Anthracene | $C_{14}H_{10}$ | 178 | 4 |
| Fluoranthene | $C_{16}H_{10}$ | 202 | 4 |
| Pyrene | $C_{16}H_{10}$ | 202 | 5 |
| Chrysene | $C_{18}H_{12}$ | 228 | 5 |
| Benzo[a]anthracene | $C_{18}H_{12}$ | 228 | 5 |
| Benzo[a]pyrene | $C_{20}H_{12}$ | 252 | 5 |
| Benzo[k]fluoranthene | $C_{20}H_{12}$ | 252 | 5 |

[a] Molecular weight (g/mol).
[b] According to van Paasen and Kiel [34].

The experimental conditions and the results of the different tests are shown in Tables 3 and 4. Before starting each test, 60 g of silica sand (or a sand/dolomite mixture, in catalysed tests) were placed in the gasifier. Once the temperature of the test was reached, the gasifier was fed with sludge and sand (20% of the mass rate of fed sludge) to improve fluidisation. In tests with catalyst, the sludge was introduced to the gasifier along with a specific sand/catalyst mixture (10/10% of the mass rate of fed sludge).

To validate each test, a mass balance closure between 95% and 105% was set, taking into account the inputs and the different products obtained.

## 3. Results and discussion

### 3.1. Influence of the TR in tests with air and without catalyst

The effect of the throughput (TR) on the produced gas composition is shown in Fig. 1a. As it can be seen, when the TR increased there was a decrease in the $H_2$ content whereas the $CH_4$, CO and

**Table 3**
Results and operating conditions of gasification experiments without dolomite.

| Parameter | Units | Test number | | | | | |
|---|---|---|---|---|---|---|---|
| | | T_1 | T_2 | T_3 | T_4 | T_5 | T_6 |
| Temperature | °C | 800 | 800 | 800 | 800 | 800 | 800 |
| Sludge | g/min | 1.2 | 2.5 | 3.7 | 1.2 | 2.5 | 3.7 |
| Sand | % fed sludge | 20 | 20 | 20 | 20 | 20 | 20 |
| Dolomite | % fed sludge | 0 | 0 | 0 | 0 | 0 | 0 |
| TR | kg/h $m^2$ | 110 | 215 | 322 | 110 | 215 | 322 |
| $u/u_{mf}$ | | 3.8 | 7.6 | 11.5 | 3.8 | 7.6 | 11.5 |
| *srt* | s | 7.5 | 3.7 | 2.5 | 7.5 | 3.7 | 2.5 |
| S/B | | | | | 1 | 1 | 1 |
| $H_2$ | %, dry | 10.3 | 9.5 | 8.5 | 11.1 | 9.7 | 8.9 |
| $N_2$ | | 59.5 | 63.6 | 63.3 | 60.6 | 63.6 | 63.3 |
| $CH_4$ | | 3.3 | 2.6 | 3.0 | 3.1 | 2.5 | 2.3 |
| CO | | 9.0 | 7.5 | 8.2 | 6.7 | 6.1 | 7.7 |
| $CO_2$ | | 13.6 | 13.2 | 13.4 | 13.7 | 13.2 | 13.9 |
| $C_2H_6$ | | 0.06 | 0.07 | 0.04 | 0.03 | 0.04 | 0.09 |
| $C_2H_4$ | | 1.9 | 2.0 | 2.1 | 1.9 | 1.9 | 2.0 |
| LHV gas | MJ/N $m^3$ | 3.6 | 3.0 | 3.1 | 3.3 | 2.9 | 2.9 |
| Gas production | N $m^3$/kg sludge, daf | 2.8 | 2.8 | 2.7 | 2.9 | 2.9 | 3.0 |
| $C_{tar}$ (gravimetric) | g/N $m^3$ | 4.1 | 7.7 | 9.8 | 3.4 | 6.0 | 7.4 |
| $Y_{tar}$ (gravimetric) | mg/g sludge, daf | 11.3 | 21.4 | 26.5 | 9.6 | 17.2 | 22.6 |
| GMB | % | 100.1 | 97.8 | 96.7 | 99.8 | 97.0 | 95.6 |
| $X_c$ | % | 73.1 | 68.1 | 69.9 | 81.0 | 66.0 | 77.5 |
| CGE | % | 43.2 | 36.1 | 36.8 | 44.9 | 35.7 | 37.8 |
| Char[a] | g/kg daf | 44.9 | 43.6 | 48.5 | 31.4 | 31.7 | 59.9 |

[a] The char content was determined according to Rapagná et al. [23].

**Table 4**
Results and operating conditions of gasification experiments with dolomite.

| Parameter | Units | Test number | | | | | |
|---|---|---|---|---|---|---|---|
| | | T_7 | T_8 | T_9 | T_10 | T_11 | T_12 |
| Temperature | °C | 800 | 800 | 800 | 800 | 800 | 800 |
| Sludge | g/min | 1.2 | 2.5 | 3.7 | 1.2 | 2.5 | 3.7 |
| Sand | % fed sludge | 10 | 10 | 10 | 10 | 10 | 10 |
| Dolomite | % fed sludge | 10 | 10 | 10 | 10 | 10 | 10 |
| TR | kg/h $m^2$ | 110 | 215 | 322 | 110 | 215 | 322 |
| $u/u_{mf}$ | | 3.8 | 7.6 | 11.5 | 3.8 | 7.6 | 11.5 |
| *srt* | s | 7.5 | 3.7 | 2.5 | 7.5 | 3.7 | 2.5 |
| S/B | | | | | 1 | 1 | 1 |
| $H_2$ | %, dry | 12.7 | 10.7 | 9.6 | 14.0 | 12.1 | 10.1 |
| $N_2$ | | 58.5 | 62.5 | 63.3 | 60.2 | 62.1 | 62.3 |
| $CH_4$ | | 3.2 | 2.6 | 2.5 | 2.4 | 2.2 | 2.8 |
| CO | | 9.7 | 6.8 | 6.8 | 5.3 | 5.4 | 6.1 |
| $CO_2$ | | 12.6 | 13.9 | 14.2 | 15.0 | 15.1 | 14.9 |
| $C_2H_6$ | | 0.04 | 0.08 | 0.09 | 0.04 | 0.04 | 0.10 |
| $C_2H_4$ | | 1.7 | 1.8 | 1.9 | 1.3 | 1.6 | 2.2 |
| LHV gas | MJ/N $m^3$ | 3.9 | 3.1 | 2.9 | 3.1 | 2.9 | 3.0 |
| Gas production | N $m^3$/kg sludge, daf | 2.9 | 2.8 | 2.8 | 3.2 | 3.0 | 3.0 |
| $C_{tar}$ (gravimetric) | g/N $m^3$ | 2.1 | 4.7 | 6.3 | 1.2 | 2.7 | 3.7 |
| $Y_{tar}$ (gravimetric) | mg/g sludge, daf | 6.0 | 13.1 | 17.4 | 3.7 | 8.1 | 11.0 |
| GMB | % | 97.5 | 97.0 | 97.5 | 95.8 | 96.7 | 95.1 |
| $X_c$ | % | 76.7 | 68.5 | 68.7 | 73.6 | 70.2 | 75.8 |
| CGE | % | 48.8 | 37.1 | 35.0 | 43.0 | 37.0 | 38.8 |
| Char[a] | g/kg daf | 47.6 | 35.7 | 30.0 | 58.1 | 38.3 | 51.1 |

[a] The char content was determined according to Rapagná et al. [23].

$CO_2$ production slightly changed. According to Chen et al. [35], the longer the *srt* (lower TR), the higher is the cracking reaction, which would partially explain the increase in $H_2$ content and the decrease in light hydrocarbons ($C_nH_m$) production obtained at lower TRs (Table 3, test 1 to test 3). Regarding the cold gas efficiency (CGE) and the carbon conversion ($X_C$), both parameters slightly decreased with higher TRs (Fig. 1b).

On the contrary, Fig. 2a shows how the TR had a large influence on gravimetric tar production. In tests without dolomite and with air as gasifying agent, tar concentration increased from 4.1 g/N $m^3$ at TR = 110 kg/h $m^2$ to 9.8 g/N $m^3$ at TR = 322 kg/h $m^2$, namely 140%. According to these results, lower *srt* (higher TRs) clearly reduce the effect of the cracking and reforming reactions on the gravimetric tar removal.

### 3.2. Influence of the steam

The results obtained in tests with air + steam (S/B = 1) and without catalyst are presented in Table 3 (test 4 to test 6). The addition of steam increased $H_2$ production due to the water–gas-shift, steam reforming and water–gas reactions [36]. At the same time, the occurrence of these reactions explains the increase of the $CO_2$

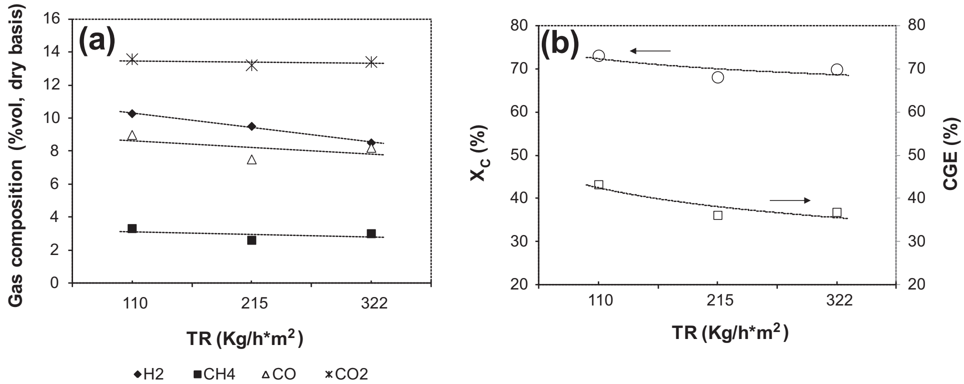


**Fig. 1.** Effect of TR on gasification products in tests without catalyst and with air: (a) gas composition (b) CGE and $X_C$.

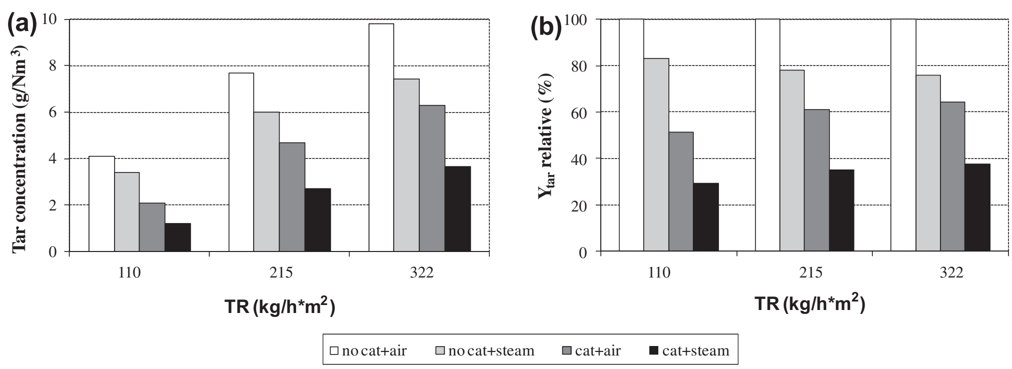


**Fig. 2.** Effect of the different gasification conditions on gravimetric tar formation: (a) tar concentration (b) $Y_{tar}$ relative (%), relation between tar production obtained in each test and tar production in test with no catalyst and air (used as reference, 100%).

production and the slight decrease of the $CH_4$ and $C_nH_m$ content. In addition, the fact that the CO content was lower than in tests with air indicates the enhancement of the water–gas-shift reaction [21,36,37].

Fig. 3 shows the produced gas composition in tests with air + steam and without catalyst under the different TRs studied. It can be seen that the $CO_2$ and $CH_4$ content scarcely varied while the $H_2$ and the CO production behaved in different ways, being the production of $H_2$ favoured by lower TRs while that of CO was impaired.

Regarding gravimetric tar concentration, Fig. 2a shows that the presence of steam decreased tar concentration in comparison to the tests with air. Specifically, the tar removal associated to the steam reforming reactions ranged between 17% and 24% (Fig. 2b).

### *3.3. Influence of the dolomite and the combined use of dolomite and steam*

Table 4 shows the results of the test with dolomite and air (test 7 to test 9) and the tests combining the use of dolomite and air + steam (test 10 to test 12). Although the use of dolomite in tests with air affected the produced gas composition (increasing the $H_2$ content) and reduced gravimetric tar concentration (Fig. 2), the most important changes were found when dolomite was used in the presence of air + steam.

For the tests with dolomite and air + steam, the $H_2$ and $CO_2$ contents increased while the CO, $CH_4$ and $C_nH_m$ production was reduced, in comparison to the tests without catalyst and with air. These results are in agreement with previous works [21,38] and may be explained mainly by the steam reforming and the water–gas-shift reactions. Additionally, the use of steam and dolomite increased the $H_2$/CO ratio from 1.1 to 2.6 at TR = 110 kg/h m$^2$ and from 1.0 to 1.7 at TR = 322 kg/h m$^2$.

Regarding the effect of TR on the gas composition in tests with catalyst, two different trends were found.

When dolomite was used with air, the $H_2$, CO and $CH_4$ content increased at lower TRs while the $CO_2$ content decreased (Table 4). According to Hernández et al. [39], lower TRs (corresponding to higher *srt*) increase the concentration of the combustible species in the produced gas as a result of the approximation to equilibrium values. On the other hand, when dolomite and air + steam were used, lower TRs increased the $H_2$ content and decreased the CO production while the $CO_2$ and $CH_4$ content slightly changed (Fig. 3), which means that the effect of the water–gas-shift reaction on the gas composition increased with the *srt*.

As far as the gravimetric tar production was concerned, the use of dolomite decreased the tar formation in comparison to test without catalyst and with air due to the reforming reactions (Fig. 2a). In fact, when dolomite was used with air, the tar removal efficiency ranged between 48% at TR = 110 kg/h m$^2$ and 36% at TR = 322 kg/h m$^2$ (Fig. 2b). Higher reductions of tar concentration were found in tests with dolomite and air + steam, with tar removal efficiencies between 71% at TR = 110 kg/h m$^2$ and 63% at TR = 322 kg/h m$^2$. It should be noted that the dolomite performance remained fairly constant over the range of TRs studied when steam was used.

### *3.4. Effect of the different gasification conditions on tar composition*

The effect of the different gasification conditions tested on the tar nature is shown in Table 5 and Fig. 4. As previously mentioned,

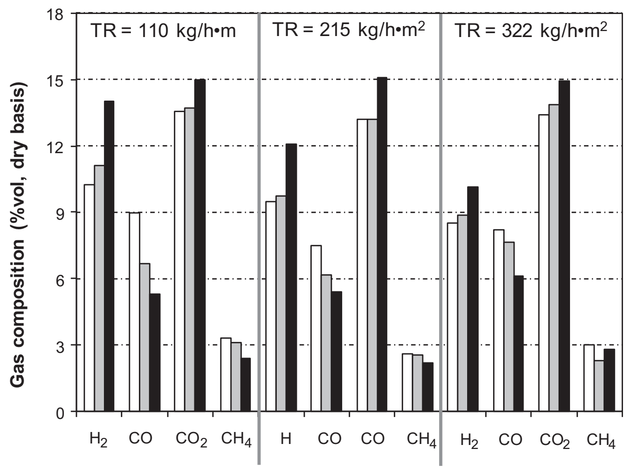


**Fig. 3.** Produced gas composition under different TRs and gasification conditions. Tests without dolomite and with air □; tests without dolomite and with air + steam ▒; tests with dolomite and air + steam ■.

the tar compounds detected by GC/MS were grouped according to the ECN classification (Table 2), distinguishing between tar class 2 (water soluble tars), tar class 3 (light aromatic hydrocarbons), tar class 4 (2–3 rings polyaromatic hydrocarbons) and tar class 5 (heavy polyaromatic hydrocarbons). Tar class 1 (GC-undetectable tars) was not estimated in this work. Among the analysed compounds, benzene was the one with the highest yield (Table 5). However, taking into account that this compound is not important for condensation and water solubility issues [34], it was removed from further analyses for a better comprehension of the results.

Fig. 4a shows the production of the different tar classes (class 2 to class 5) obtained under different gasification conditions. The results have been divided in four groups: tests with no catalyst and air (no cat + air), tests with no catalyst and air + steam (cat + steam), tests with dolomite and air (cat + air) and tests with dolomite and air + steam (cat + steam). Fig. 4b presents this information but in terms of relative tar composition.

As it can be seen in Fig. 4a, the effect of the TR on the GC-detectable tar was not so evident than on the gravimetric tar (Fig. 2a). In all the tests, the gravimetric tar production clearly increased with higher TRs. However, GC-detectable tar production shown different trends depending on the tar class analysed. In tests without catalyst, the total GC-detectable tar behaved like the gravimetric tar and the tar production increased with TR. On the other hand, in tests with dolomite, the maximum GC-detectable tar production was found at medium TR (215 kg/h $m^2$). Taking a look to Fig. 4b, it can be seen that the relative content of tar class 2 decreased with lower TRs (higher *srt*) while tar class 4 content increased. According to [34], higher *srt* changes the tar composition from mainly light compounds to a mixture of heavy and light tars. These heavy tars would correspond to tar class 4 (or even tar class 5) produced by the dimerisation of aromatic compounds or by cyclisation or polymerisation reactions of unsaturated C2—C4 hydrocarbons, which was in agreement with the lower $C_2H_4$ content obtained at lower TRs (Tables 3 and 4).

Deepening in the analysis of the different tar classes, Fig. 4a shows that the use of steam slightly increased tar class 2 (water soluble tars) production. On the contrary, there was a relevant decrease when dolomite was used, both in tests with air and with air + steam. According to these results, the use of primary dolomite could be an interesting option together with water scrubbing systems downwards the gasifier to reduce hazardous waste water production and thus waste disposal cost [34].

As far as tar class 3 (light aromatic hydrocarbons) was concerned, the tar production increased only when dolomite was used with air and no significant changes were found with any of the other primary measures tested (Fig. 4a). According to Fig. 4b, tar class 3 is the prevailing class (corresponding to 40% of the GC-

**Table 5**
GC-detectable tar production and gas dew point.

| | T_1 | T_2 | T_3 | T_4 | T_5 | T_6 | T_7 | T_8 | T_9 | T_10 | T_11 | T_12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| *Tar production (mg/g sludge, daf)* | | | | | | | | | | | | |
| Benzene | 26.9 | 21.2 | 20.0 | 30.7 | 21.7 | 20.5 | 39.4 | 24.6 | 15.4 | 30.3 | 20.9 | 10.1 |
| Tar class – 2 | 7.5 | 10.8 | 11.5 | 9.2 | 11.4 | 14.2 | 5.0 | 7.4 | 5.0 | 3.1 | 7.7 | 6.3 |
| Tar class – 3 | 8.8 | 13.6 | 12.9 | 11.8 | 14.5 | 11.0 | 10.7 | 18.1 | 14.6 | 9.2 | 13.3 | 9.4 |
| Tar class – 4 | 7.4 | 8.2 | 9.0 | 7.6 | 7.4 | 7.5 | 5.7 | 5.2 | 5.2 | 4.5 | 4.8 | 3.4 |
| Tar class – 5 | 0.27 | 0.49 | 0.63 | 0.24 | 0.40 | 0.52 | 0.11 | 0.16 | 0.17 | 0.04 | 0.19 | 0.18 |
| Gas dew point (°C) | 144.0 | 144.9 | 155.4 | 144.1 | 148.6 | 150.7 | 132.8 | 143.4 | 143.5 | 110.9 | 131.9 | 156.5 |

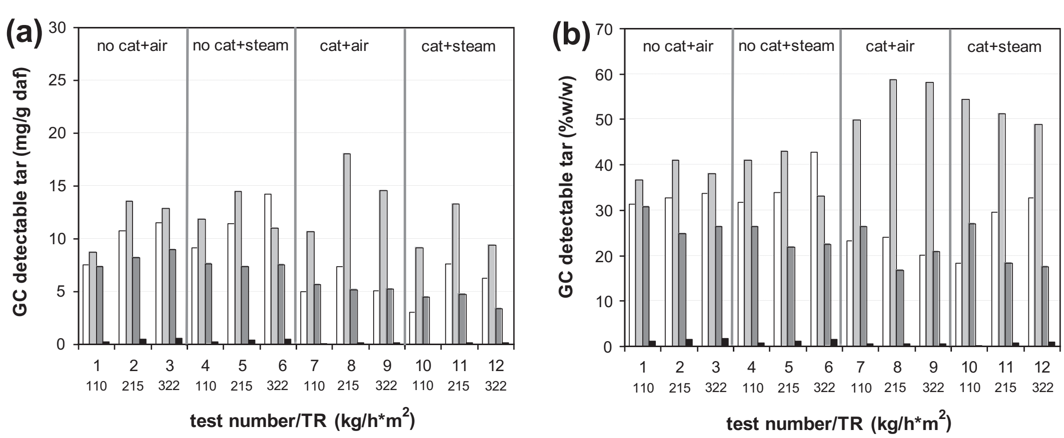


**Fig. 4.** Effect of the different gasification conditions on tar production by tar classes: (a) tar production (b) tar weight distribution. Tar class 2 □; Tar class 3 ; Tar class 4 ▒; Tar class 5 ■.

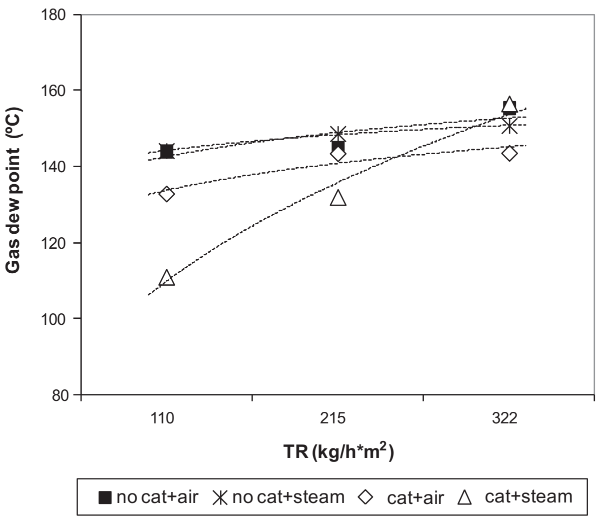


**Fig. 5.** Effect of the different gasification conditions on the gas dew point.

detectable tar production) in tests without catalyst. Moreover, the use of dolomite increased the relative importance of this class, reaching relative values between 50-60%. These results are in agreement with those obtained with other primary additives (alumina and lime) by Campoy et al. [9].

Regarding tar class 4 (2–3 rings polyaromatic hydrocarbons), Fig. 4a shows that the use of steam slightly decreased tar production in comparison to tests with air. However, the use of dolomite, and specially dolomite and air + steam, clearly reduced the production of these compounds. Similar results were found in terms of relative tar composition (Fig. 4b). In fact, the activity of dolomite in tar class 4 conversion (taking naphthalene as representative) has been previously demonstrated [40].

As for tar class 5 (heavy polyaromatic hydrocarbons), it can be seen in Fig. 4a that steam had a slight influence over this tar class while dolomite effectively removed these heavy compounds. Even though the production and the relative importance of tar class 5 is much lower than that of the rest of the classes (represents less than 2% of the tar content, Fig. 4b), these compounds determine to a large extent the dew point of the produced gas, limiting the final use of the gas when it has to be cooled before use [9].

Fig. 5 shows the effect of the different gasification conditions on the gas dew point. In tests without catalyst and with air, the effect of the TR on this parameter was limited and variations around 10 °C were found over the range of TR tested. In addition, the dew point hardly changed when steam was added.

Higher reductions where found with dolomite, especially at low and medium TRs (110 and 215 kg/h $m^2$, respectively) and mainly when catalyst and steam were used. These gasification conditions corresponded to the highest reductions of tar class 4 and tar class 5 (Fig. 4).

Regardless of the above mentioned, the gas dew point never dropped below 110 °C (Table 5), a temperature far above the recommended 30 °C for syngas engine applications [11].

## 4. Conclusions

Some tests were carried out to study the effect of throughput (TR), steam, and the use of dolomite (as primary catalyst) on sewage sludge gasification products. The analysis of the results was mainly focussed on the effect of these primary measures on gas composition, tar production and composition.

Even though TR slightly affected the cold gas efficiency and the carbon conversion, higher TRs decreased the $H_2$ content of the produced gas and increased the gravimetric tar production. Higher TRs correspond to lower gas residence times, reducing the effect of the cracking and reforming reactions on the gravimetric tar removal.

In comparison to the tests with air, the use of air + steam (S/B = 1) as gasifying agent increased the $H_2$ and $CO_2$ content and decreased the CO production. Moreover, the gravimetric tar yield fell around 21% due to the promotion of the steam reforming reactions.

The use of dolomite had positive effects on the gasification products, especially when it was used with air + steam. In comparison with tests without catalyst, the $H_2$ content increased between 12% and 26%. Regarding gravimetric tar production, reductions of up to 71% were reached with the combined use of dolomite and steam in comparison to tests with air and without catalyst. Dolomite performance remained fairly constant over the range of TRs studied.

The effect of TR on the GC-detectable tar was different depending on the tar class analysed. Lower TRs (higher residence times) increased the relative presence of tar class 4 and decreased that of tar class 2. Tar class 5 production increased with higher TRs while tar class 3 seemed to be favored by medium TR (215 kg/h $m^2$).

From a practical point of view, the use of primary dolomite decreased the production of tar class 2 (water soluble tar) so this catalyst could be an interesting option in combination with water scrubbing systems downwards the gasifier to reduce waste disposal costs.

Under the experimental conditions tested, the gas dew point never dropped below 110 °C, value far above the recommended temperature when the syngas is to be used for engine applications. Considering this, technical problems can be expected related to tar condensation if additional tar removal techniques are not implemented.

These results try to provide enough insight on the potential extension of this process to a larger scale. However, this extrapolation might not be straightforward as the scale and the particularities of the process limit the suitability of the present results. For example, it could happen that sewage sludge is not milled to a small size therefore affecting the gasification rate due to limitations in heat and mass transfer imposed by particle size and thus the obtained products. An additional variation to be expected is the fact that industrial TRs would be higher than those tested. Finally, the feasibility of the tested operation conditions needs to be contrasted with those occurring in typical autothermal applications.

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## Glossary

*daf:* Dry and ash-free
*ER:* Equivalence ratio, defined as the ratio between the flow rate of air introduced into the gasifier and the stoichiometric flow rate of air required for complete combustion of the sludge
*LHV:* Low heating value of the produced gas, MJ/N $m^3$, dry basis
*SB:* Steam-to-biomass ratio, defined as the flow rate of steam fed to the reactor divided by the flow rate of sludge (daf)
*u:* Superficial gas velocity in the gasifier bed, cm/s
$u_{mf}$: Minimum fluidisation gas velocity (gasifier bed conditions), cm/s
*N* $m^3$: Cubic metre, normal conditions (0 °C, 101 kPa)
$Y_{gas}$: Gas yield, N $m^3$ dry gas/kg sludge, daf
$C_{tar}$: Gravimetric tar concentration, g/N $m^3$
$Y_{tar}$: Gravimetric tar yield, mg/g sludge, daf
*GMB:* Global mass balance, %
$X_c$: Carbon conversion, weight of carbon in the produced gas divided by weight of carbon in the sludge introduced in the gasifier
*CGE (cold gas efficiency):* LHV of gas divided by the LHV of sludge